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This program of research was established to develop CdTe epitaxial films and quantum well structures by molecular beam epitaxy (MBE) and to study their physical properties. During 1983-1985, CdTe films were successfully deposited on various substrates including sapphire, silicon, and gallium arsenide. The films were judged to be of high quality on the basis of photoluminescence and x-ray diffraction rocking curve studies.

In 1983, the first CdTe quantum well structures in the world were grown at NCSU under NSF support. These multilayers were composed of CdMnTe barrier layers alternating with CdTe quantum wells. They also represented the first quantum structures ever grown which contained layers of a diluted magnetic semiconductor (CdMnTe). Teh CdMnTe-CdTe superlattices proved to be intense sources of luminescence, particularly at low temperatures. Stimulated emission from CdTe quantum wells was first observed at NCSU in 1984. In 1985, the first demonstrations of stimulated emission from a CdMnTe laser structure was also observed at NCSU. CdMnTe lasers grown by MBE at NCSU were also studied at the National Magnet Laboratory at MIT in 1986 where magnetic tuning of the emission wavelength was demonstrated and attributed to the exchange interaction in DMS materials.

In order to control the electrical properties of CdTe and related materials by substitutional doping, a new deposition technique, photoassisted MBE, was developed at NCSU. This technique uses a laser to illuminate the substrate surface during film growth. Using this new approach, n-type and p-type films of CdTe were successfully propared using In and Sb as dopants, respectively. Subsequently, As was also used as a p-type dopant. More recently, the technique has been extended to study doping in CdMnTe as well as HgCdTe, an important infrared detector material.

Using the photoassisted MBE technique to prepare n-type CdTe and CdMnTe layers led to the fabrication of field effect transistors based on these materials.

The present program has impacted profoundly on the development of the II-VI semiconductors studied and has contributed significantly to a strong resurgence of interest in these materials. The work impacts on the areas of optoelectronics, magnetooptics, integrated optics and infrared detector technology.

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FINAL REPORT

ARO Contract DAAG29-84-K-0039 (1984 - 1988)

Title: SYNTHESIS AND PROPERTIES OF CdTe FILMS

A. Summary of Important Results

MBE Growth of CdTe Films on Alternative Substrates. During 1984-1985, CdTe films were deposited by MBE on various alternative substrates including sapphire [1], silicon [2], indium antimonide [3], and gallium arsenide [4]. The best CdTe heteroepitaxy was obtained on basal plane sapphire, (100) GaAs, and (111)B GaAs. In each of these cases, double crystal x-ray diffraction rocking curves as narrow as 60 arc sec were obtained, indicating a high degree of structural perfection had been obtained. Photoluminescence studies at low temperatures were completed and yielded spectra dominated by sharp near-edge exciton peaks. Defect band luminescence at 1.40 - 1.46 eV was generally absent from the high quality epitaxial layers [5-8]. Heteroepitaxial CdTe films were subsequently used as high resistivity substrates for the growth of HgCdTe films and CdMnTe-CdTe superlattices at NCSU.

Growth of CdMnTe-CdTe Multilayer Structures and Superlattices. The first CdTe quantum well structures in the world were grown at NCSU in 1983 [9]. These multilayers were composed of CdMnTe barrier layers alternating with CdTe quantum wells. These initial superlattice structures were grown on CdTe/sapphire substrates. They also represented the first quantum structures ever grown which contained layers of a diluted magnetic semiconductor (CdMnTe).

The CdMnTe-CdTe superlattices proved to be intense sources of luminescence, particularly at low temperatures [10]. In addition, new insights concerning the nature of excitons in DMS layered structures were obtained from luminescence studies performed in high magnetic fields at the National Magnet Laboratory at MIT [11,12]. In particular, it was found that a portion of the luminescence from the DMS layered structures was highly polarized, indicating that the excitons in the CdTe quantum wells were localized near the CdMnTe interfaces where the magnetic exchange interaction is appreciable.

Stimulated emission from CdTe quantum wells was first observed at NCSU in 1984 [13]. In these experiments, a pulsed argon ion laser was used to optically pump cleaved cavities composed of CdMnTe-CdTe superlattice layers on GaAs. Subsequently, in 1985, the first demonstration of stimulated emission from a CdMnTe laser structure was also observed at NCSU [14]. CdMnTe laser structures grown at NCSU were also studied in 1986 at the National Magnet

Laboratory where magnetically tunable stimulated emission was observed [15]. The tunablility range of the emission was found to be much larger (by a factor of fifty) than for that of non-magnetic semiconductors. This is again due to the magnetic exchange interaction in DMS materials which occurs between Mn ions and the conduction band electrons and valence band holes. This interaction gives rise to large positive electronic g-factors so that the band gap of a DMS semiconductor changes appreciably under an applied magnetic field. Work in this area is continuing in collaboration with Dr. Peter Wolff and others at the MIT facility.

It should be pointed out that all of the initial work involving CdTe quantum wells and other DMS layered structures involved samples that were all found to be electrically insulating. Our initial attempts to dope CdTe layers were not very successful, due to compensation effects, as has also plagued the development of n-type and p-type bulk CdTe. To circumvent this fundamental problem required the development of a new approach to doping--photoassisted MBE--that was initiated at NCSU beginning in 1985.

Photoassisted MBE: A New Approach to Epitaxy and Doping. At NCSU, under NSF support, we have developed a new technique for substitutional doping of II-VI semiconductor films, photoassisted MBE, in which the substrate is illuminated during the deposition process as shown in Fig. 1. In our initial studies [16-22] an argon ion laser operating with broad band yellow-green optics was used as an illumination source. However, we have recently shown that conventional (incoherent) light sources can also be used in certain applications. The laser power density at the substrate during film deposition is approximately 150 mW/cm^2 which produces a negligible (≤ 2 °C) temperature increase.

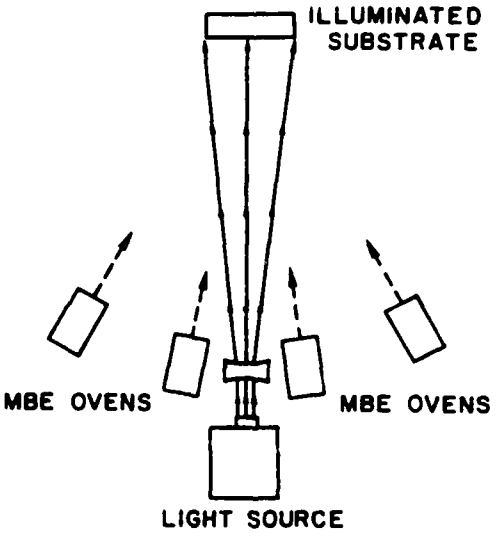

Fig. 1. Photoassisted molecular beam epitaxy film growth process.

The rationale behind the photassisted MBE technique is to provide high energy, low momentum particles (photons) at the film growth surface which can influence surface chemical reactions during the deposition process. Effects which may result from the introduction of light include enhancement of the surface mobility of atoms, modification of surface bonds, conversion of surface molecules into atoms, and modification of the electrical potential of the surface through generation of photoexcited carriers. On the basis of recent experiments, it appears that surface photochemical reactions may play a major role in the changes in epitaxy that occur because of the photoassist.

Illumination of the substrate during the growth process was found to cause a profound change in the electrical properties of CdTe epilayers. Only those films grown under illumination were conducting. Films grown by conventional MBE are semi-insulating. In contrast to this, undoped CdTe films grown by photoassisted MBE are n-type and exhibit high electron mobilities and low carrier concentrations at low temperatures. This behavior is illustrated by the Hall data shown in Figs. 2 and 3 for three different films grown by photoassisted MBE. Note that electron mobilities as large as 6,600 $cm^2/V \cdot s$ have been achieved. To our knowledge, this is the largest electron mobility ever achieved for a CdTe film grown by any technique. In addition, the undoped films show a high degree of structural perfection as manifested by very narrow x-ray rocking curve FWHMs (Fig. 3).

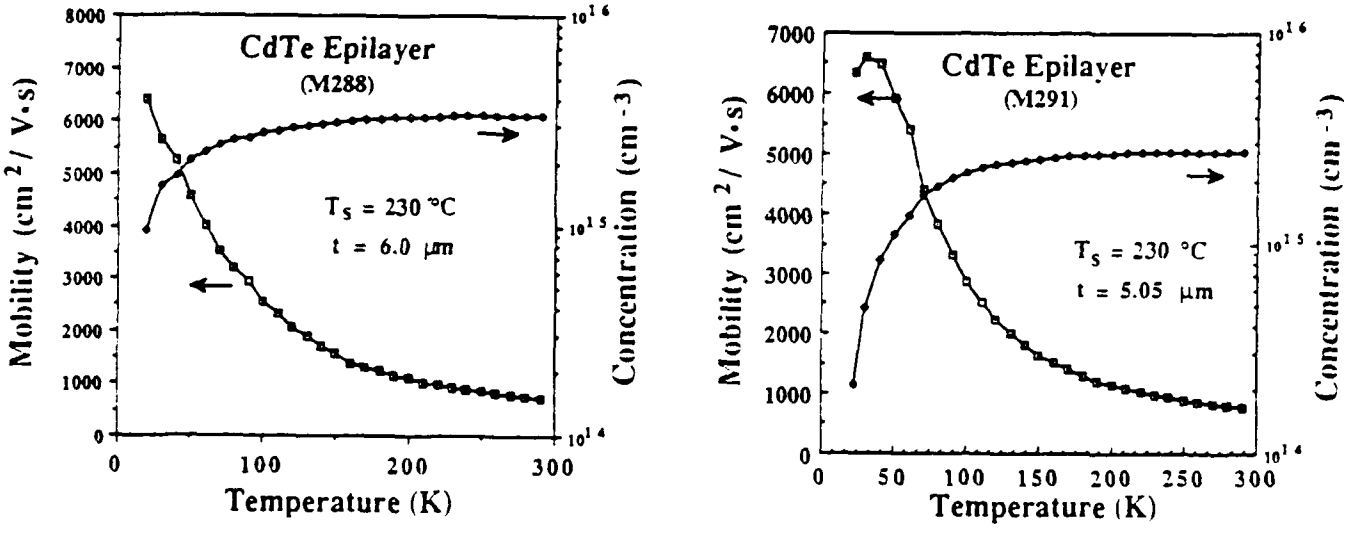

Fig. 2. Hall effect data for two CdTe films grown by photoassisted MBE.

Both n-type and p-type substitutionally doped CdTe films have been successfully grown using the photoassisted MBE technique. The best n-type doping results were obtained for CdTe:In samples grown on high quality CdTe substrates at 230 °C. Room temperature mobilities ranged

from 450-800 $cm^2/V \cdot s$ and increased with decreasing temperature. An example of this behavior is shown in Fig. 4 in which Hall data for a CdTe:In epilayer is shown. The highest mobility of 2,400 $cm^2/V \cdot s$ occurs at T = 80 K.

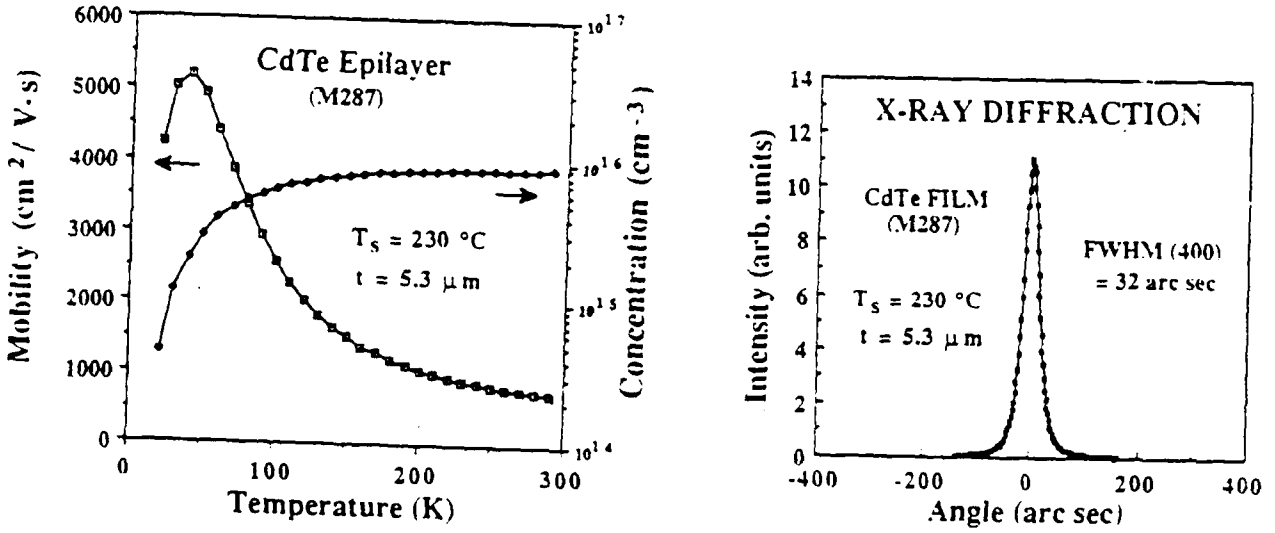

Fig. 3. Hall effect and x-ray diffraction rocking curve data for CdTe film M287.

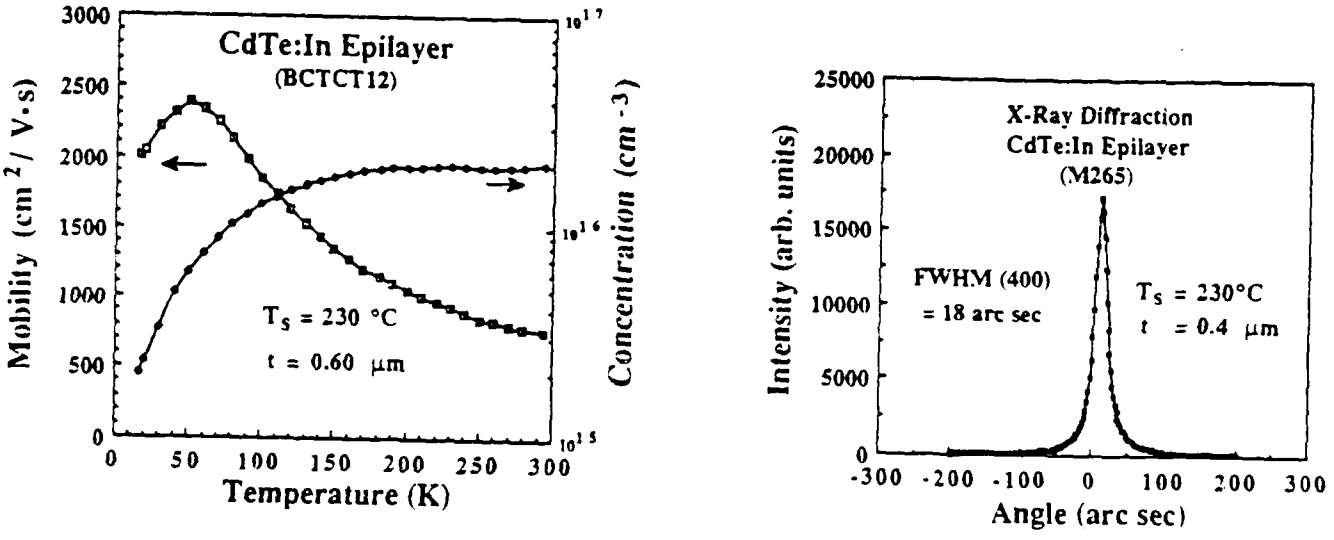

Fig. 4. Hall effect and x-ray diffraction data for CdTe:In films.

P-type CdTe films have been successfully prepared by photoassisted MBE using Sb [17] and As as dopant species. In both cases, p-type layers having hole concentrations ranging from 10^{16} to greater than 2×10^{18} cm^{-3} at 300 K have been grown. Hole mobilities at room temperature range from 50 to 82 $cm^2/V \cdot s$. Fig. 5 shows photoluminescence (PL) and Hall effect data which

illustrate several important properties of the doped CdTe:Sb and CdTe:As films, respectively. As seen from the figure, the luminescence from CdTe:Sb is very bright and consists principally of a single sharp line at about 1.5894 eV, which is identified as an acceptor-bound exciton line. Other features of the PL spectrum are due to excited states of the bound excition and a phonon replica at 1.5683 eV. In addition, the feature at 1.5414 eV is an electron-to-acceptor transition. Assuming this is the Sb acceptor, and using the band gap of CdTe (1.606 eV), we obtain 64.6 meV as the ionization energy for the Sb acceptor in CdTe. Similar PL spectra for CdTe:As films yield 58 meV for the As acceptor level. The Hall data shown at the right of Fig. 5 is for a CdTe:As film. Note that the hole mobility at 300 K is 74 $cm^2/V \cdot s$ and increases to 105 $cm^2/V \cdot s$ at 240 K. These mobilities are comparable to hole mobilities in the best bulk p-type CdTe. In addition, it is extremely important to note the high dopant concentration of 6×10^{18} cm^{-3} that has been achieved for CdTe:As films grown by the photoassisted MBE technique. Similarly high dopant concentrations up to 3×10^{18} cm^{-3} have also been obtained for p-type CdTe:Sb films. These values are nearly two orders of magnitude greater that hole concentrations achieved via substitutional doping of bulk CdTe crystals. This strongly suggests to us that non-equilibrium photochemical reactions at the growth surface play a role in the photoassisted MBE process such that incorporation of active p-type dopants is greatly enhanced.

Hall effect measurements for p-type CdTe samples at temperatures below 300 K are difficult to complete because of problems in obtaining ohmic contacts to this p-type material. At present, we are investigating this contacting problem in detail at NCSU and have recently made

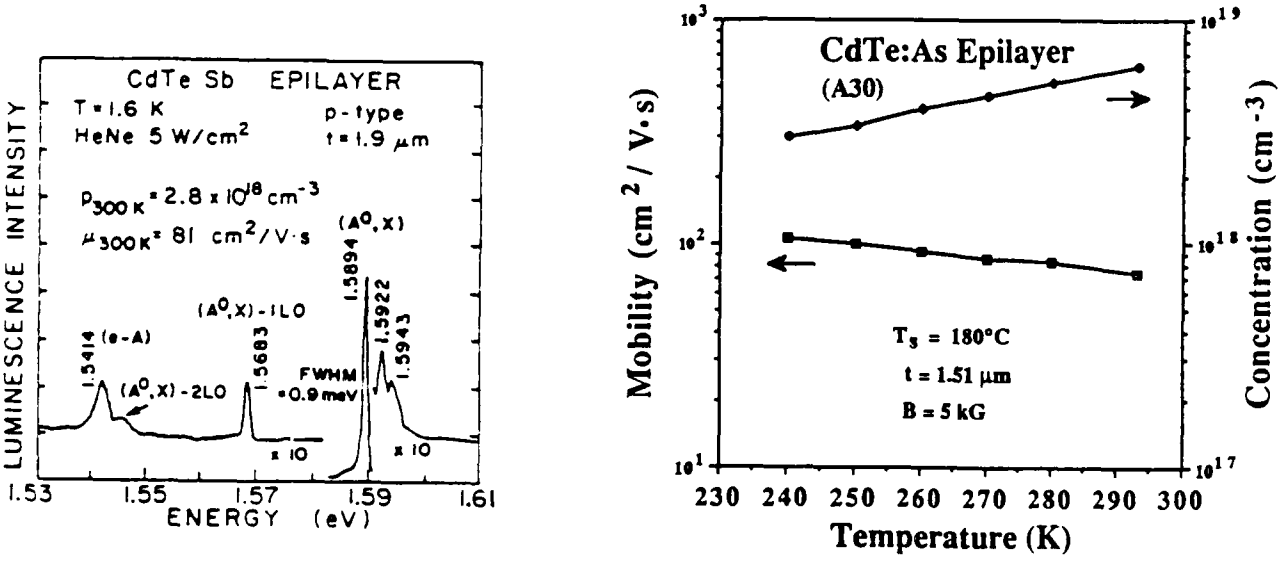

Fig. 5. Photoluminescence and Hall effect data for p-type CdTe films.

considerable progress. Thus, we are confident that multilayer structures, such as double heterostructures or superlattices, which might require p-type CdTe with low resistance ohmic contacts, can now be fabricated at NCSU.

Doped CdMnTe-CdTe Superlattices. The first successful substitutional doping of diluted magnetic semiconductor superlattices composed of alternating layers of CdMnTe:In and CdTe has been accomplished at NCSU using the photoassisted MBE technique. In order to achieve this result it was first necessary to learn to dope CdMnTe films with indium. This was accomplished in 1986 when films of CdMnTe:In having electron concentrations in excess of 10^{17} cm^{-3} were prepared. Since then, a number of conducting CdMnTe:In-CdTe superlattices have been prepared and studied by means of Hall effect [21], photoluminescence [22], Raman scattering [23], and x-ray diffraction experiments [24]. These studies have provided new and fundamental information concerning the role of dopants in DMS structures. In particular, dopant ionization energies in CdMnTe have been measured from PL and Hall effect data, excited states of quantum well structures have been determined by means of excitation PL studies and, more recently, piezoreflectance studies, and the nature of electronic wavefunctions in CdTe quantum wells have been probed via spin-flip Raman experiments using CdMnTe-CdTe superlattices having CdTe quantum wells of different thicknesses. Because of the exchange interaction which is responsible for the spin-flip Raman line, these types of experiments have also provided information concerning wafe-function penetration into CdMnTe DMS barrier layers. These studies are continuing at the present time.

Fabrication and Testing of II-VI Semiconductor Devices. Because of our success in controlling the electrical properties of CdTe and CdMnTe films, we have been able to initiate a program at NCSU to fabricate and test electronic devices based on these materials. In particular, p-n junctions of CdTe have been prepared by photoassisted MBE by depositing CdTe:Sb onto a CdTe:In epitaxial film. Mesa structures were then fabricated by means of photolithography and I-V characteristics were measured and reported [19]. These diode structures represent an important advancement in the development of this II-VI compound semiconductor since they represent the first all-thin-film CdTe diodes prepared by any technique and, more importantly, because n-type and p-type CdTe layers can serve as injecting layers in a double heterostructure (DH) device such as CdTe:In-HgCdTe:CdTe:Sb. The development of DH devices of this type will be a major goal of the research to be described in Section III of this proposal, which will emphasize the development of wide-band-gap HgCdTe ($x > 0.3$) for use as sources and detectors in the 1.0 - 5.5 μm spectral range.

More recently at NCSU, we have fabricated the first metal-semiconductor field effect transistors (MESFET's) using both CdTe:In [25] and CdMnTe:In conducting epitaxial layers grown by photoassisted MBE. Fig. 6 shows a schematic of the transistor mesa structure employed along with source, drain, and gate electrodes. At the right are source-to-drain I-V curves for different reverse-bias gate voltages applied to a depletion-mode FET. Note that excellent depletion mode transistor action has been achieved with pinch-off voltages of about 1.5 V. For other FET's having 5 μm wide gates, pinch off voltages of 4 V and transconductances of 10 mS/mm were obtained for devices having a channel doping density of about 10^{17} electrons /cm^3. Additional FET's, including the first DMS transistor which employs a doped CdMnTe:In epilayer, have also recently been successfully fabricated and studied at NCSU [26].

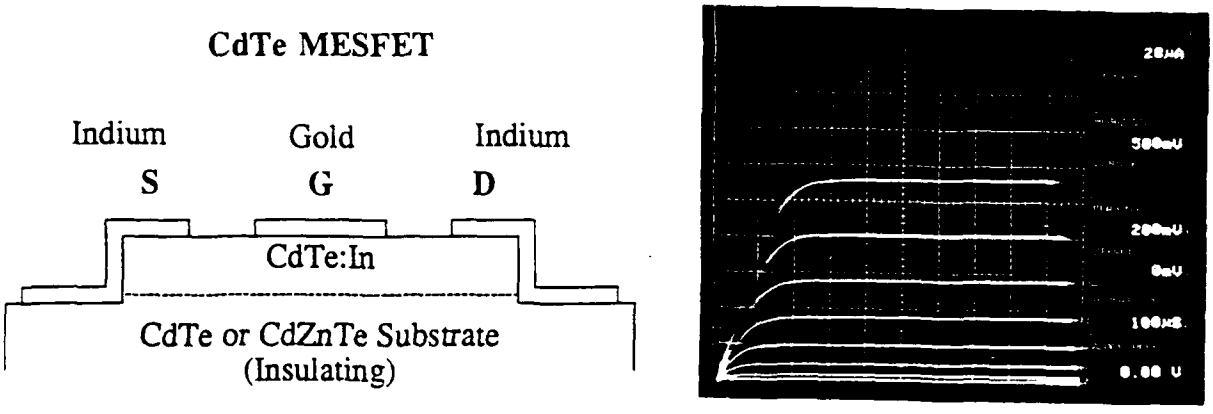

Fig. 6. CdTe MESFET structure and measured transistor I-V curves.

Summary. During the current NSF grant (1983 - present) considerable progress has been made in preparing and studying high quality CdTe and CdMnTe thin films and quantum well structures prepared by MBE and photoassisted MBE. This work, along with additional work completed concurrently under DOD support that deals with MBE-growth of small-band-gap HgCdTe ($x = 0.2$) films which will be summarized later, provides a firm basis of accomplishments for the research to be described in Section III of this proposal.

B. Publications Resulting from ARO Support (1983 - 1988)

PUBLICATIONS IN REFEREED JOURNALS

1. *"Growth of CdTe Films on Sapphire by Molecular Beam Epitaxy"*, T.H. Myers, Yawcheng Lo, R.N. Bicknell, and J.F. Schetzina, Appl. Phys. Lett. **42,** 247 (1983).

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12. *"Properties of $Cd_{1-x}Mn_xTe$-CdTe Superlattices Grown by Molecular Beam Epitaxy"*, R.N. Bicknell, N.C. Giles-Taylor, D.K. Blanks, R.W. Yanka, E.L. Buckland, and J.F. Schetzina, J. Vac. Sci. Technol. **B 3,** 709 (1985).

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22. *"Strain Effects in $Cd_{1-x}Mn_xTe$-CdTe Superlattices"*, D.K. Blanks, R.N. Bicknell, N.C. Giles-Taylor, J.F. Schetzina, A. Petrou, and J. Warnock, J. Vac. Sci. Technol. **B 4**, 635 (1986).

23. *"Growth and Properties of Dilute Magnetic Semiconductor Superlattices Containing $Hg_{1-x}Mn_xTe$"*, K.A. Harris, S. Hwang, Y. Lansari, J.W. Cook, Jr. and J.F. Schetzina, Appl. Phys. Lett. **49**, 713 (1986).

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25. *"Stimulated Emission from $Cd_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$-$Cd_{1-y}Mn_yTe$ Multiple Quantum Well Structures"*, R.N. Bicknell, N.C. Giles-Taylor, N.G. Anderson, W.D. Laidig, and J.F. Schetzina, J. Vac. Sci. Technol. **A 4**, 2126 (1986).

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29. *"Stimulated Emission from $Cd_{1-x}Mn_xTe$-CdTe and $Cd_{1-x}Mn_xTe$-$Cd_{1-y}Mn_yTe$ Multiple Quantum Well Structures"*, R.N. Bicknell, N.C. Giles-Taylor, J.F. Schetzina, N.G. Anderson, and W.D. Laidig, Proc. of the 1986 Seoul International Symposium on the Physics of Semiconductors and Its Applications, Seoul National University, Seoul, Korea, pp. 149 (1986).

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32. *"Growth and Properties of Dilute Magnetic Semiconductor Superlattices Containing $Hg_{1-x}Mn_xTe$"*, K.A. Harris, S. Hwang, Y. Lansari, R.P. Burns, J.W. Cook, Jr. and J.F. Schetzina, J. Vac. Sci. Technol. **B 5**, 699 (1987).

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INVITED PAPERS AND COLLOQUIA

J.F. Schetzina, Principal Investigator

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2. *"Properties and Appplications of CdTe/Sapphire and CdTe/GaAs Thin Films"*, Duke University (1983).

3. *"Growth and Properties of CdTe Films on Alternative Substrates"*, General Electric Corporate Research Laboratory, Schenectady, NY (1983).

4. *"MBE Growth of CdTe/Sapphire and CdTe/GaAs Films"*, Ford Aerospace and Commmunications Laboratory, Newport Beach, CA (1983).

5. *"Properties of CdMnTe-CdTe Superlattices"*, Symposium on Diluted Magnetic Semiconductors, MIT National Magnet Laboratory , Cambridge, MA (1984).

6. *"Properties of Superlattices Composed of II-VI Semiconductor Compounds"*, American Chemical Society Meeting, Raleigh, NC (1984).

7. *"MBE Growth of Novel II-VI Semiconductor Films and Multilayered Structures"*, University of Maryland (1984).

8. *"Quantum Well Structures and Superlattices Composed of II-VI Semiconductors Containing Magnetic Ions"*, American Physical Society March Meeting, Baltimore (1985); BAPS **30**, 298 (1985).

9. *"Growth of Novel II-VI Semiconductor Films and Quantum Well Structures by Molecular Beam Epitaxy"*, University of South Carolina (1985).

10. *"Growth and Properties Of Novel Quantum Well Structures Containing Magnetic Ions"*, IBM Research Laboratory, San Jose (1985).

11. *"DMS Quantum Well Structures"*, MIT National Magnet Laboratory (1985).

12. *"Molecular Beam Epitaxy of II-VI Semiconductors"*, Ford Aerospace Laboratory, Newport Beach, CA (1985).

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14. *"Dilute Magnetic Semiconductor Quantum Wells"*, Purdue Unversity (1986).

15. *"Properties of II-VI Quantum Well Structures Grown by MBE"*, University of Notre Dame (1986).

16. *"Quantum Well Structures Containing Magnetic Ions"*, Martin Marietta Laboratories, Baltimore (1986).

17. *"Submicron Heterostructures of DMS Materials"*, Purdue University (1986).

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19. *"MBE Growth of II-VI Films, Quantum Well Structures, and Superlattices"*, Texas Instruments Central Laboratory, Dallas (1986).

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D.K. Blanks, Post Doctoral Research Asssociate

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R.N. Bicknell, Ph.D. Student

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35. *"Growth and Characterization of High Quality, Low Defect, Subgrain Free Cadmium Telluride by a Modified Horizontal Bridgman Technique"*, W.P. Allred, A.A. Khan, C.J. Johnson, N.C. Giles, and J.F. Schetzina, MRS Meeting, Boston, MA (1986).

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39. *"Properties of Doped II-VI Films and Superlattices Grown by Photoassisted MBE"*, N.C. Giles, R.N. Bicknell, and J.F. Schetzina, BAPS **32**, 600 (1987).

40. *"Non-Meanfield Spin-Glass Behavior of Bulk CdMnTe"*, T. Datta, A. Barrientos, J. Amirzadeh, E.R. Jones, Jr., and J.F. Schetzina, BAPS **32**, 630 (1987).

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42. *"Properties of Substitutionally Doped CdMnTe Films and CdMnTe-CdTe Quantum Well Structures"*, R.L. Harper, S. Hwang, N.C. Giles, R.N. Bicknell, and J.F. Schetzina, 8th MBE Workshop, Los Angeles, CA (1987).

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45. *"Properties of Doped CdTe Films Grown by Photoassisted Molecular Beam Epitaxy"*, S. Hwang, R.L. Harper, K.A. Harris, N.C. Giles, R.N. Bicknell, J.W. Cook, Jr., and J.F. Schetzina, 1987 MCT Workshop, New Orleans, LA (1987).

46. *"Electrical Properties of CdTe Metal-Semiconductor Field Effect Transistors"*, D.L. Dreifus, R.M. Kolbas, J.R. Tassitino, R.L. Harper, R.N. Bicknell, and J.F. Schetzina, 1987 MCT Workshop, New Orleans, LA (1987).

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48. *"Structural Properties of II-VI Semiconductor Bulk Substrates, Thin Films, and Superlattices"*, J.K. Gobble, K.A. Bowers, J.N. Matthews, S. Hwang, R.L. Harper, and J.F. Schetzina, BAPS **32**, 2137 (1987).

C. Student Awards Received for Work Supported by ARO

Nancy Catherine Giles, Ph.D. Student at NCSU, recipient of the inaugural Louise Meyer Schutzmeister Award in 1983 given annually by the Association of American Women in Science to the most outstanding female physics graduate student in the U.S.A., awarded Ph.D. in Solid State Physics by NCSU in 1987.

Robert Newton Bicknell, Ph.D. Student at NCSU, recipient of a National Student Award given by the Materials Research Society in 1986 for his pioneering work leading to the development of diluted magnetic semiconductor superlattices and laser structures, Ph.D. in Solid State Physics awarded by NCSU in 1986.

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